Discerning different nucleation mechanisms in synthetic trachybasalts: example from three Titanomagnetite populations in single experiments

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Heterogeneous nucleation, i.e. nucleation on a pre-existing surface, is energetically more favourable than homogeneous nucleation because it requires overcoming a lower energetic barrier in order to form a critical nucleus. In most natural and experimental crystallizing magmas, heterogeneous nucleation is suggested to be the main nucleation mechanism, but robust criteria to prove the heterogenous nucleation origin of a given crystal or phase in natural rocks and ex-situ samples are lacking.

Here we apply multiple analytical methods in order to assess the nucleation mechanism of titanomagnetite (Tmt) crystals that crystallized in the proximity of clinopyroxene (Cpx) crystals in a synthetic trachybasaltic melt (with 2 wt.% added H_2O) in crystallisation experiments carried out in a piston-cylinder apparatus at a constant pressure of 4 kbar. After 30 minutes of superheating at 1300 °C, the samples were cooled at a rate of 80°C / min to the final resting temperatures of 1150 °C and 1100 °C. These temperatures correspond to a respective undercooling (ΔT expressed as T liquidus – T experiment) of 30° and 80°. The dwell times at these temperatures were 30 minutes and 8 hours, respectively.

3D image processing and 3D image analysis of high-resolution synchrotron X-ray computed microtomography (SR μCT) data resulted in a precise phase segmentation of Cpx, Tmt, glass and bubbles. Moreover, it was possible to discriminate three main Tmt populations spatially distributed alongside Cpx crystals but morphologically different to each other: a) Tmt grains > 100 μm in size, skeletal in shape, and mostly isolated in the melt (population 1); b) Tmt grains between up to 100 μm in size, anhedral to partially skeletal in shape, and always decorating Cpx grains edges and tips (population 2); c) needle-to-flattened Tmt grains almost completely enclosed within Cpx grains (population 3).

The pair correlation function g(r), i.e. a measure of the frequency of an interpoint distance (of r), has been evaluated for the 3D point pattern composed by the centroids of each Tmt population, extracted from several VOIs inside 3D scans of the samples. Tmt grains of population 1 have an unclear 3D point pattern characterized by g(r) near 1, a possible sign of a randomly distributed point pattern. Populations 2 and 3 show clear clustered 3D point patterns, characterized by g(r) > 1 and interpoint distances r up to 200 μm.

Electron backscatter diffraction (EBSD) analysis enables us to clarify the crystallographic orientation relationships (CORs) between Cpx and Tmt crystals which share boundaries. Less than 60 % of the total shared Cpx-Tmt boundary length of population 1 Tmt crystals is characterized by the presence of CORs. Individual crystals of this population show no CORs between the two crystal phases at all, or boundary misorientations dispersed around known specific CORs. In contrast, more than 85% of the total shared Cpx-Tmt boundary length in Tmt populations 2 and 3 follows a COR. Locally, clusters of multiple Tmt decorating single Cpx crystals show more than 95% of the cumulative shared length characterized by the presence of CORs.

Considering the skeletal-to-euhedral shape, the unclear 3D point pattern distribution and the lack of CORs in some Cpx-Tmt pairs classified as Population 1, we suggest that the isolated single grains from this class are most likely formed by homogeneous nucleation. Individual large grains of this same population showing CORs may represent unusually large heterogeneously nucleated Tmt grains, Cpx grains heterogeneously grown on a pre-existing Tmt crystal, or potentially Tmt-Cpx interaction after nucleation apart.

Considering the anhedral or acicular shape, the highly clustered point pattern, and the ubiquitous presence of CORs, we interpret Population 2 and 3 Tmt grains to have formed by heterogeneous nucleation on top of pre-existing Cpx crystals.

In conclusion, multiple Tmt morphologies and distributions coupled with different COR systematics imply different nucleation mechanisms and growth histories for the three populations. Notably, the different Tmt nucleation mechanisms occurred during or right after a single cooling event. Multiple microstructural populations of crystals in natural magmas should be carefully assessed before inferring the existence of complex thermal (or other) histories. A multi-methodological approach which combines 3D SR μCT data with 2D crystallographic one is indispensable to confidently discern between homogeneous and heterogenous nucleation mechanisms.

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